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A simulation model for methane emissions from landfills with interaction of vegetation and cover soil

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ABSTRACT

Global climate change and ecological problems brought about by greenhouse gas effect have become a severe threat to humanity in the 21st century. Vegetation plays an important role in methane (CH_4) transport, oxidation and emissions from municipal solid waste (MSW) landfills as it modifies the physical and chemical properties of the cover soil, and transports CH_4 to the atmosphere directly via their conduits, which are mainly aerenchymatous structures. In this study, a novel 2-D simulation CH_4 emission model was established, based on an interactive mechanism of cover soil and vegetation, to model CH_4 transport, oxidation and emissions in landfill cover soil. Results of the simulation model showed that the distribution of CH_4 concentration and emission fluxes displayed a significant difference between vegetated and non-vegetated areas. CH_4 emission flux was 1–2 orders of magnitude higher than bare areas in simulation conditions. Vegetation play a negative role in CH_4 emissions from landfill cover soil due to the strong CH_4 transport capacity even though vegetation also promotes CH_4 oxidation via changing properties of cover soil and emitting O_2 via root system. The model will be proposed to allow decision makers to reconsider the actual CH_4 emission from vegetated and non-vegetated covered landfills.

1. Introduction

Landfilling remains the primary treatment option for municipal solid waste (MSW) and other non-hazardous wastes in most parts of the world. Landfill gas (LFG, mainly CH_4 and CO_2) are produced during the stabilization process of MSW. Among of them, CH_4 is the most concerned as the second potent greenhouse gas (GHG) after CO_2 with a 21 times higher global warming potential than that of CO_2 over a 100-year period (IPCC, 2013). Following agriculture and coal mines, landfills are the third largest anthropogenic CH_4 emission sources. They were reported to emit about 138 million metric tons CO_2 equivalent ($MMTCO_2e$) in 2015 and accounted for about 17.7% of the USA CH_4 emissions (Chai et al., 2016). In 2030, emissions from landfills are expected to represent 10% of the global total methane from all sources (EPA, 2013). CH_4 is not only a greenhouse gas but also an energy reservoir, as it can be reused as a clean fuel or combusted for power generation or heat supply if an appropriate LFG collection system is in place (Aydi et al., 2015). Unfortunately, less than 50% of landfills have installed LFG collection system in China (Chai et al., 2016), and the collection efficiency varies from 8% to 90%, depending on the cover type (daily, intermediate and finally covers, with or without geomembrane), collection well type (horizontal or vertical well) and coverage (Börjesson et al., 2007; De la Cruz et al., 2016; Goldsmith et al., 2012; Spokas et al., 2006; Sun et al., 2015; Zhang et al., 2010). Consequently, a large amount of fugitive CH_4 is emitted to the atmosphere through daily and intermediate cover soil without geomembrane covered.

Due to the uncertainties and complexities association with the CH_4 production, consumption and transport process, the CH_4 emission flux in field landfills ranged over seven orders of magnitude from less than 0.0004 $mg\,m^{-2}\,d^{-1}$to more than 10,000 $mg\,m^{-2}\,d^{-1}$(Spokas et al., 2006). In some case, landfill can even be a sink for atmospheric CH_4 (Schuetz et al., 2003; Spokas et al., 2006). Compared with direct or indirectly measurement methods such as flux chamber, differential adsorption LiDAR, tracer dilution methods and stochastic search method (Abichou et al., 2006; Babilotte et al., 2010; Geck et al., 2016; Kormi et al., 2016; Taylor et al., 2016), the method of model, i.e., Intergovernmental Panel on Climate Change (IPCC) model, California Air Resources Board (CARB) model to predict CH_4 emissions from landfills is primarily applied due to the technological and economical limitation (De la Cruz et al., 2016). A few models have been established to assess the CH_4 transport and oxidation in landfill cover soil. Bogner et al. (1997) built a dynamic model for LFG transport and CH_4

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oxidation in landfill cover soils based on the mechanism of diffusion and convection. The model is attractive and innovative, but the CH_4 oxidation is modelled via the growth of bacteria on the surface of soil sphere by empirical equation instead of the factors influencing the growth of bacteria such as the water content, porosity and organic matters, thus the further validation of the model is limited for an unreliable predictive capacity for CH_4 oxidation in landfill cover soil. Some researchers used Fick's law and CH_4 oxidation kinetics to simulate CH_4 diffusion and oxidation in landfill soils (Bogner et al., 1997; Spokas et al., 2011; Stein et al., 2001). As the concentration of CO_2 in LFG can be as high as 40–50%(v/v) (Schuetz et al., 2003), Fick's law becomes unsuitable to analyze the LFG transport process as it only used in (i) binary mixtures, (ii) diffusion of dilute species in a multicomponent mixture (Webb and Pruess, 2003), and (iii) in the absence of electrostatic or centrifugal force field (Krishna and Wesselingh, 1997; Vural et al., 2010). Instead, the Maxwell-Stefan equation, a gas mixture diffusion equation, is more suitable to describe the LFG transport in landfill cover soils. De Visscher and Van Cleemput (2003) and Hilger et al. (1999) used the Maxwell-Stefan equation to simulate the LFG transport process. In the model of Hilger et al. (1999), O_2 is the only limiting substrate as the depth of methanotrophically active zone is limited by O_2 penetration, but it may lead to overestimate of CH_4 oxidation close to soil surface, where the concentration of CH_4 is low. The diffusion coefficients vary with the soil properties and climate conditions, the model built by Stein et al., 2001 and De Visscher and Van Cleemput (2003) applied concentration-dependent diffusion coefficients which improved the reliability and accuracy. Diffusion is usually viewed as the dominant transport mechanism in cover soil and convection can be negligible. However, continuous generation of LFG from the waste layer, temporal and spatial variability of meteorological conditions, and properties of the cover soil will nevertheless induce pressurization in the cover soil layer, and convection may thus also plays an important role in CH_4 transport(Park et al., 2016; Rannaud et al., 2009; Yao et al., 2015). A better understanding of CH_4 transport, oxidation and emission from landfill cover soils is needed to take proper measures to mitigate CH_4 emissions into the atmosphere.

After a landfill site has been closed, the surface of the cover soil will be covered by native plants (Xiaoli et al., 2011). Plants in the cover soil may have a positive influence on CH_4 oxidation as plants are reported to emit O_2 to the cover soil by spreading roots (Bohn et al., 2011; Colmer, 2003). Vegetation also alters the chemical and physical properties of the cover soil such as the density, moisture content and soil porosity. Furthermore, plant growth may provide nutrients and carbon source for methanotrophs by root exudates and debris of dead plants, thus improving CH_4 oxidation capacity (Bohn et al., 2011). However, certain plant species especially vascular plants may have a negative impact on CH_4 emission, as CH_4 may be released directly to the atmosphere through the aerenchyma (Rusch and Rennenberg, 1998).

The impact of vegetation on CH_4 transport, oxidation and emission has been modelled in rice paddies and wetlands (Li et al., 2016; Xu et al., 2007). The importance of vegetation in landfills, however, is not yet incorporated in landfill models. The CALMIM (California Landfill Methane Inventory Model) that incorporates both site-specific soil properties and soil microclimates based on 1-D diffusion is the most comprehensive model and validated in field landfills. In this model, vegetation coverage was included, but it mainly used to modify incoming solar radiation (Spokas et al., 2011); Abichou et al. (2015) modelled the effect of vegetation on CH_4 transport, oxidation and emissions in landfill across different climates. Their results suggest that the impact of vegetation on CH_4 oxidation and emission mainly works by changing the soil characteristics. They nevertheless neglected to incorporate the direct CH_4 transport by vegetation leaf stomata and stems (Rusch and Rennenberg, 1998).

The aim of this study is to develop a new simulation model that combines the multicomponent diffusive equation, convective equation and the dual Monod kinetic equation coupled with an embedded vegetation module to clarify the mechanisms of CH_4 transport, oxidation and emission in landfill soils covered with vegetation. The result of the simulation model will be proposed to allow decision makers to reconsider the actual CH_4 emission from vegetated and non-vegetated covered landfills.

2. Model description

The landfill cover soil is an unsaturated porous media. The LFG produced in the process of waste degradation is emitted to the atmosphere through the porous media by diffusion and convection, at the same time, part of CH_4 is oxidized during the process of transport by methanotrophs in the cover soil. The LFG transport and CH_4 oxidation are considered as the dominant processes governing CH_4 emission from the landfill cover soil. The multicomponent diffusive equation, Darcy's law and the dual Monod kinetic equation are the basic equations involved in the model (Fig. 1).

2.1. LFG diffusion model

Molecular diffusion of multicomponent gas mixture in unsaturated porous media is governed by the Maxwell-Stefan diffusive equation in landfill cover soil:

$$\frac{-P}{RT}\frac{\partial y_i}{\partial z} = \sum_{\substack{j=1 \\ k\neq i}}^{n} \frac{N_i y_k - N_k y_i}{D_{soil,ik}} \tag{1}$$

with $D_{soil,ij}$ the binary diffusion coefficient of a mixture of gases i and j in soil matrix ($m^2\ s^{-1}$), z the depth (z = 0 m at the soil surface), y_i the mole fraction of component i, P the absolute pressure (Pa), R the universal gas constant (8.3145 J $mol^{-1}\ K^{-1}$), N_i the flux of component i, i and k present the gas type (1-CH_4, 2-CO_2, 3-O_2, 4-N_2).

We used the Wilke approximation, a method of using Fick's law with variable diffusivities to solve Maxwell-Stefan diffusion:

$$j_i = -\left(\rho y_i \sum_k D_{soil,ik} d_k\right) \tag{2}$$

where j_i is the average flux of gas i (kg $m^{-2}\ s^{-1}$); ρ is the density of gas mixture (kg m^{-3}); d_k is the diffusive driving force of gas i (m^{-1}).

When the landfill gasses are in ideal state, then:

$$d_k = \nabla x_k + \frac{1}{P_A}[(x_k - y_k)\nabla P_A] \tag{3}$$

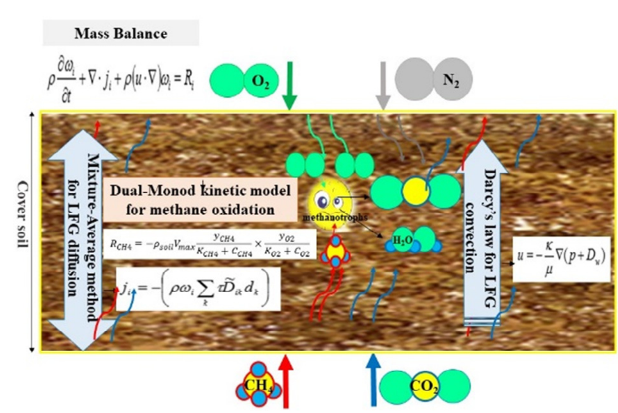

Fig. 1. The schematic diagram of the model.

$$x_k = \frac{y_k}{M_k} M_n \tag{4}$$

$$M_n = \left\{ \sum_i \frac{y_i}{M_i} \right\}^{-1} \tag{5}$$

where x_k is the molar fraction of gas k; P_A is the air pressure (Pa); M_i is the molar mass of gas i (kg mol^{-1}); M_n is the mean molar mass of the gas mixture (kg mol^{-1}). $D_{soil,ik}$ can be calculated according to Millington-Quirk equation (Millington and Quirk, 1961):

$$D_{soil,ik} = D_{air,ik} \frac{\varepsilon^{\frac{10}{3}}}{(\varepsilon + \omega)^2} \tag{6}$$

with ε the air-filled porosity and ω is the water content. $D_{air,ik}$ can be calculated by the Eq. (7) according to the diffusion coefficient D_{ik}^T in the multicomponent diffusive equation (Stein et al., 2001):

$$D_{air,ik} = \frac{1 - y_i}{\sum_{k=1, i \neq k}^{m} \frac{y_i}{D_{ik}}} \tag{7}$$

$D_{air,ik}^T$ is calculated by Eq. (8) (Spokas et al., 2011):

$$D_{air,ik}^T = D_{air,ik}^{293.15} \left(\frac{T}{293.15} \right)^{1.75} \tag{8}$$

with $D_{air,ik}^{293.15}$ the binary diffusion coefficient in air at 293.15 K (m^2 s^{-1}) and T is the temperature (K).

2.2. LFG convection model

We use Darcy's law to describe the process of convective transport of LFG in the cover soil. The equation of Darcy's law and mass balance are as follows:

$$u = -\frac{\kappa_e}{\mu} \nabla P_A \tag{9}$$

$$\frac{\partial y_i}{\partial t}(\rho\varepsilon) + \nabla \cdot (\rho u) = Q_m \tag{10}$$

where u is the constant velocity of soil gas transport upwards in steady state (m s^{-1}); κ_e is the effective permeability of cover soil (m^2); μ is the gas kinematic coefficient of viscosity (Pa s); ε is the air-filled porosity (m^3 gas m^{-3} soil) and Q_m is the total mass source (kg m^{-3} s^{-1}).

2.3. Methane oxidation kinetics model

The progress of CH_4 oxidation in the landfill cover soil can be described by the dual Monod kinetic equation in porous media:

$$R_{CH4} = -\rho_{soil} M_{CH4} f_{V,T} f_{V,m} V_{max} \frac{y_{CH4}}{K_{CH4} + C_{CH4}} \times \frac{y_{O2}}{K_{O2} + C_{O2}} \tag{11}$$

where R_{CH4} is the CH_4 oxidation rate (mol kg^{-1} s^{-1}); V_{max} is the maximum CH_4 oxidation rate (mol kg^{-1} s^{-1}); K_{O2} and K_{CH4} are the half-saturation rate constants of O_2 and CH_4, respectively (dimensionless); ρ_{soil} is the density of cover soil (kg m^{-3}); M_{CH4} is the molar mass of CH_4 (kg mol^{-1}); y_{CH4} and y_{O2} are the mole fraction of CH_4 and CO_2 in the cover soil, respectively; $f_{V,T}$ and $f_{V,m}$ are the modified factor for the maximum CH_4 oxidation rate which can be obtained via empirical equation (Eqs. S(1) and S(2) in the supporting materials. CH_4 is partial oxidized to carbon dioxide and the rest is converted to biomass (—CH_2O—), which is then further converted to CO_2 (De Visscher and Van Cleemput, 2003). The chemical reaction equation can be described as follows:

$$CH_4 + O_2 = CO_2 + 2H_2O \tag{12}$$

$$R_{O_2} == 2R_{CH_4} \tag{13}$$

$$R_{CO_2} == -R_{CH_4} \tag{14}$$

where R_{O2} was the consumption rate of O_2 via CH_4 oxidation (mol kg^{-1} s^{-1}) and R_{CO2} was the production rate of CO_2 via CH_4 oxidation (mol kg^{-1} s^{-1}).

2.4. The continuity equation

The continuity equation for gas i can be written as

$$\rho \frac{\partial y_i}{\partial t} + \nabla \cdot j_i + \rho(u \cdot \nabla) y_i = R_i \tag{15}$$

$$N_i = j_i + \rho u y_i \tag{16}$$

where R_i is loss rate of gas i by biodegradation (kg m^{-3} s^{-1}) and N_i is the total emission flux of gas i (kg m^{-2} s^{-1}).

2.5. Vegetation embedded model

The vegetation embedded model consists of three modules:(1) the plant growth changes the properties of cover soil such as water content, soil porosity, the maximum CH_4 oxidation rate V_{max} and the half-saturation rate constant K_{CH4} and K_{O2}; (2) O_2 from the atmosphere or produced by photosynthesis is transported in the cover soil via the root system; (3) CH_4 can be emitted to the atmosphere by vegetation roots. No mature models have been currently established for the estimation of the radial oxygen loss (ROL) of different types of vegetation. Colmer, however, showed that ROL was positively correlated with the specific surface area and dry mass of roots (Colmer, 2002; Colmer, 2003). We hence used Eq. (17) to model the ROL (Segers and Leffelaar, 2001).

$$\mathrm{ROL} = [(55.3\beta + 0.07) \times 10^{-6} \mathrm{mol\ kg^{-1}\ s^{-1}}] M_{root} / \pi L_{root}^2 \tag{17}$$

where ROL is the radial oxygen loss (mol m^{-2} s^{-1}); M_{root} is the dry mass weight of root (g); L_{root} is the maximum root length (m) and β is the root specific surface area which is determined by the type of vegetation (Table 1).

The root system not only transports O_2 to the cover soil layer, but also transports CH_4 to the atmosphere. The equation of CH_4 emission by vegetation can be described by empirical equations as follows (Kirk, 1997; Wissuwa and Ae, 2001; Xu et al., 2007):

$$R_{vegetation} = -\eta \frac{M_{CH4} D_{rs} C_{CH4}}{V_m \varepsilon} A_z \tag{18}$$

$$A = 0.0689 M_{root}^{0.8162} (M_{root} = (0, 15]\mathrm{g}) \tag{19}$$

$$A_z = 2A / (4/3 \pi L_{root}^3) \tag{20}$$

where $R_{vegetation}$ is the CH_4 transport rate by vegetation (kg m^{-3} s^{-1}); D_{rs} is the transport velocity of CH_4 across the root epidermis (m s^{-1}); A_z is the average root surface area per unit volume of soil (m^2 m^{-3}); A is the total surface area of roots (m^2); η is the modified CH_4 transport coefficient; D_{rs} can be calculated with Eq. (21) (Segers and Leffelaar, 2001):

$$D_{rs} = K_{rt} \cdot \omega_{gas} \tag{21}$$

Table 1
The root specific surface area parameters.

Type	Woody plants	Gramineous plants	Herbaceous plants
Taproots system	1	0.4	0.2
Fibrous root system	–	0.6	0.3

where K_{rt} is the absorption rate of water by roots (m^3 H_2O m^{-2} s^{-1}) and ω_{gas} is the ratio of gas to water in the liquid absorbed via roots (m^3 gas m^{-3} water).

2.6. Model boundary conditions and assumptions

A few assumptions were made to set up the model (Table 2).

In our simulation model, CH_4 transport and oxidation are considered as the main processes involving in CH_4 emissions in the cover soil, and CH_4 production is omitted. The intermediate cover soil with a depth of 0.5 m was covered with vegetation. As shown in Fig. 1, the boundary conditions of the model are determined by geometric boundary conditions of the cover soil layer: the upper boundary conditions include atmosphere pressure and gas volume fraction of air in the cover soil surface, and the bottom boundary conditions include the CH_4 production flux in the interface of cover soil layer and waste layer. The CH_4 production flux can be measured in the field or estimated by CH_4 production models.

The module to incorporate the effect of vegetation on CH_4 transport and oxidation also follows the assumptions listed in Table 2, and additionally the following two assumptions are included: (1) The CH_4 transport occurs in the plant roots growth zone V1, a semicircular area with the center of the interface between plant and cover soil surface, with L_{root} as the radius; (2) the area V2 where the physical properties of cover soil changed by the plant growth, is a semicircular area with the center of the interface between plant and cover soil surface, with $L_{root}+\delta$ as the radius (Fig. 2).

2.7. Model application

As a partial validation, we conducted a comparison of CH_4 emission flux with field measurements. Field experiments were performed in June 2014 in the Jiangcungou landfill, which is located in Xi'an city, China (N34°14′58″, E100°06′53″). Jiangcungou landfill currently accepts 3600 tons of MSW per day. Two landfill cells with both vegetated and bare areas had been closed for 1.5 and 2 years, and were equipped with passive LFG collection systems. Sandy loam with a height of 35–40 cm was applied as the intermediate cover soil. The surface of the investigated areas was covered by 9 types of vegetation. Following phyto-sociological methods of the Braun-Blanquet School, we only considered two dominant species: *Alkali kochia* (*A. kochia*) and *Annual ryegrass* (*A. ryegrass*).

The LFG emission flux was measured using the static chamber method in both vegetated and bare areas. Two landfill cells named cell 1 and cell 2 with two vegetated areas X-V1 and X-V2 covered by *A. kochia* and *A. ryegrass* respectively, and bare areas X-B1 and X-B2 were measured over an entire day (Fig. S1). The CH_4 production flux was measured after the cover soil was dug out.

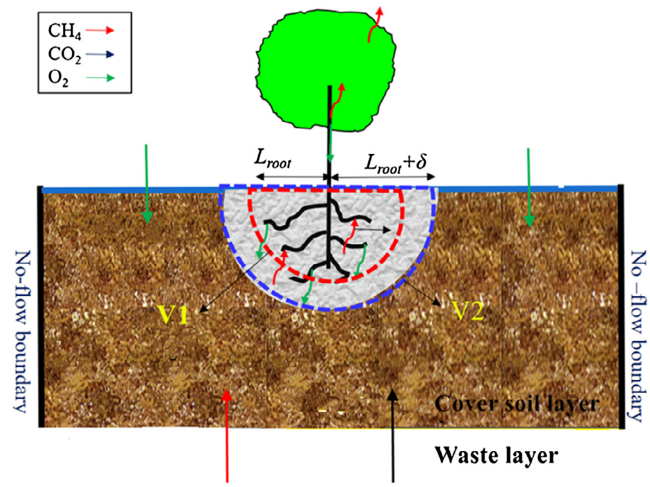

Fig. 2. The basic schematic of the model in landfill covered with vegetation.

2.8. Model simulation

Model simulations were performed using COMSOL Multiphysics software (Version 5.2a, COMSOL Co., Ltd.), a commercially available software based on the finite element method (FEM). LFG concentration distribution in vertical profile, emission flux and CH_4 oxidation rate data are used as outputs. Table 3 presents the model input parameters in vegetation areas and bare areas. The parameters of the vegetation embedded model are: L_{root} = 0.25 m, δ = 0.05 m and M_{root} = 5 g. Other parameters are listed in supporting information (Table S1). The simulation areas are free discrete into finite element unit of triangle according to the geometric characteristic automatically. The unit side length of the triangle is less than 0.02 m, to ensure that stability and symmetry of the simulation (Fig. 3).

3. Results and discussion

3.1. Sensitivity analysis

The reference of the values of the parameters are shown in Table 3. According to previous studies, the CH_4 uptake rate in landfill cover soils ranges from 1.6×10^{-6} mol m^{-2} s^{-1} to 0.0031 mol

Table 2
Assumptions employed to develop the model.

	Assumption	Reason
#1	The physical and chemical characteristics of cover soil are horizontally homogeneous	The model mainly focuses on the vertical gas transport
#2	CH_4, CO_2, O_2 and N_2 are the only gasses considered in the model	The total volume fraction of CH_4, CO_2, O_2 and N_2 in the LFG is more than 99% (De Visscher and Van Cleemput, 2003), other gasses such as aromatic compounds, chlorinated hydrocarbons, organic sulfur compounds, H_2S, and N_2O may also exist in the LFG, but their volume fraction is less than 2%
#3	The anaerobic CH_4 oxidation in the cover soil is omitted; O_2 is the only electron acceptor in the CH_4 oxidation progress	Though anaerobic CH_4 oxidation has been reported in many natural environments, the contribution ratio is very low as the slower oxidation rate, (Knittel and Boetius, 2009; Scheutz et al., 2009), so the anaerobic CH_4 oxidation can be omitted
#4	Soil respiration is omitted	The CO_2 produced by respiration accounts for 1–3% (Bogner et al., 1997), so soil respiration in this study is negligible
#5	The dissolution of CO_2 in cover soil is omitted	Although solubility of CO_2 is much higher than CH_4, it has little influence on LFG transport (De Visscher and Van Cleemput, 2003)
#6	The palnt root-water uptake is omitted	Even though reduction of moisture by plant root-water uptake could improve soil aeration for microbial aerobic CH_4 oxidation in landfill cover soils, excessive soil water removal could suppress microbial activity due to water shortage (Feng et al., 2017). We only consider the gas transport and oxidation in the cover soil, thus the palnt root-water uptake can be omitted

Table 3
Model input parameters for the simulation conditions.

Parameter	ω	ε	L	$F_{in\text{-}CH4}$	V_{max}	K_{CH4}	K_{O2}
Unit			m	mol $m^{-2}s^{-1}$	mol kg^{-1} s^{-1}		
Vegetation areas	0.15	0.5	0.5	1×10^{-4}	1.0×10^{-7}	0.0091	0.0200
Bare areas	0.2	0.3	0.5	1×10^{-4}	7.3×10^{-7}	0.0066	0.0120
Sources	Default value				Modified according to De Visscher and Van Cleemput (2003)		

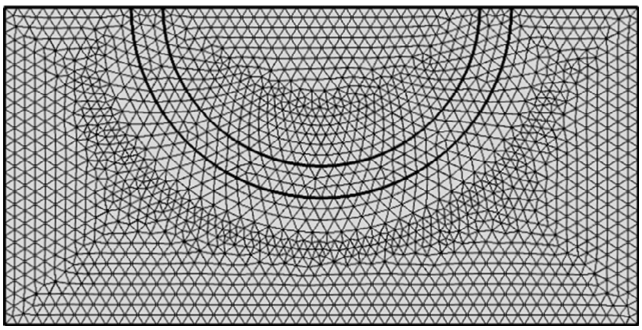

Fig. 3. The distribution of mesh in the simulated landfill cover soil areas.

m^{-2} s^{-1} (De Visscher et al., 1999; De Visscher and Van Cleemput, 2003; Gabarro et al., 2013; Yao et al., 2015). Therefore, we selected five CH_4 production rates ($F_{in\text{-}CH4}$ = 10^{-3}, 10^{-4}, 10^{-5}, 5×10^{-5} and 10^{-6} mol m^{-2} s^{-1}) for the sensitivity analysis. Sensitivity analysis for V_{max} ranges from 1×10^{-8} to1 $\times 10^{-6}$ mol kg^{-1} s^{-1} based on the reported values from the literature (Abichou et al., 2009; Park et al., 2005; De Visscher et al., 1999). Other parameters are varied from 20% to 200% of their reference values in Table 3 for the bare areas. Other weather conditions are setting as default value (Table S1). The sensitivity of the model to the main parameters are shown in Fig. 4. Obviously, the predicted CH_4 oxidation efficiency are more sensitive to the parameters of V_{max}, $F_{in\text{-}CH4}$, ε and K_{CH4}. Thus, choose reasonable values of the parameters are key factors to improve the accuracy of predication.

3.2. Vertical profile of soil gas concentration distribution in cover soil

Fig. 5 shows the simulated concentration of soil gas (CH_4, CO_2 and O_2) and the emission fluxes, using measured and default values of parameters (N_2 concentration is presented in supporting information Fig. S2). Due to consumption of CH_4 by methanotrophs, the concentration of CH_4 decreased from the bottom to the surface (De Visscher and Van Cleemput, 2003; Stein et al., 2001; Yao et al., 2015). The volume fraction of CH_4 and CO_2 in the bottom layer was 28% and 29% (v/v), respectively, which was lower than the input values (60% and 40%, v/v, respectively) as the N_2 diffusion (Fig. S2) in the cover soil. The decay rate of CH_4 was much higher than for CO_2 in the 0.25–0.5 m depth layer, which suggests that CH_4 is more easily emitted to the surface, and CO_2 on the contrast is more likely to accumulate in the bottom layer (Spokas et al., 2015). Influenced by the plant root systems, the high soil porosity and the effective diffusion coefficient, concave shapes of CH_4 and CO_2 concentration distribution occurred in the cover soil. As a result, CH_4 decay rate and CO_2 production rate significantly increased when gas was transported by vegetation. The gradient of CH_4 concentration was steeper where bare and vegetated areas meet. CH_4 concentration was below 0.05% in the vegetated areas. The penetrative depth of O_2 in vegetated areas can be as deep as 0.2 m (De Visscher and Van Cleemput, 2003; De Visscher et al., 1999), which is much deeper than in bare areas. At the same time, the O_2 diffusion flux was much higher in vegetated area than in bared area. CH_4 oxidation is hence enhanced in vegetated areas by O_2 permeation.

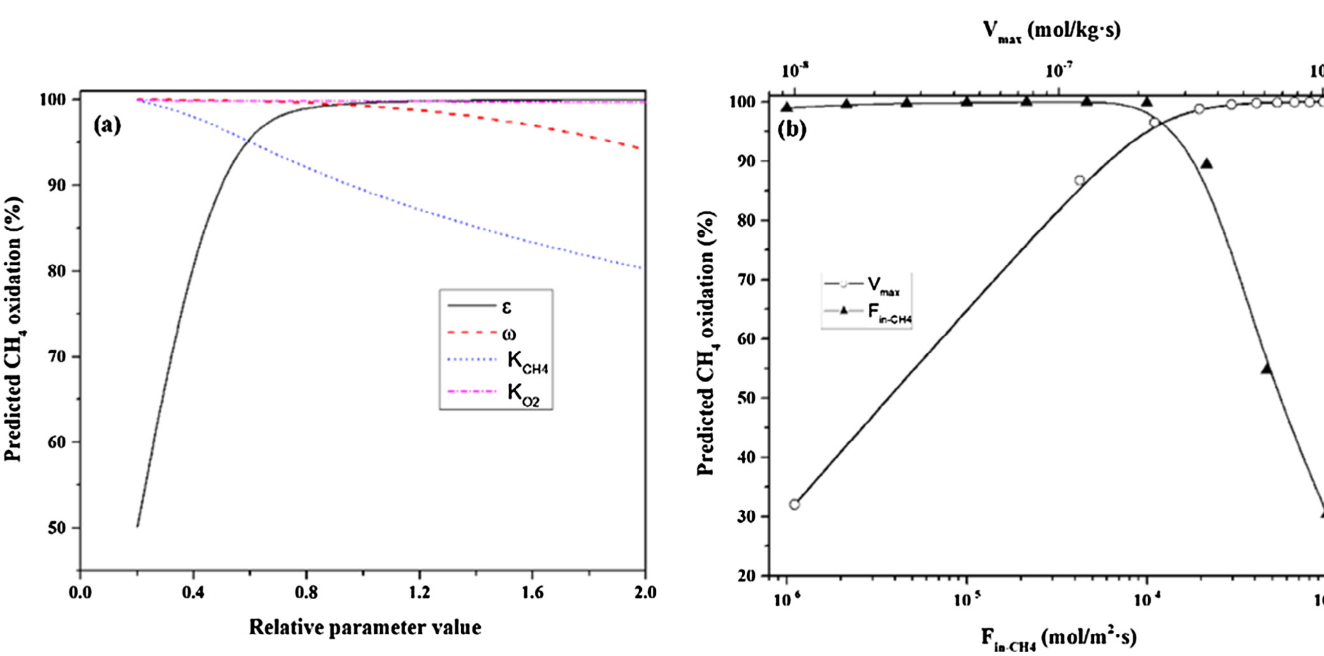

Fig. 4. Influence of model parameters on CH_4 oxidation. X-axis of (a) is parameter value divided by reference value of Table 3 in bare areas.

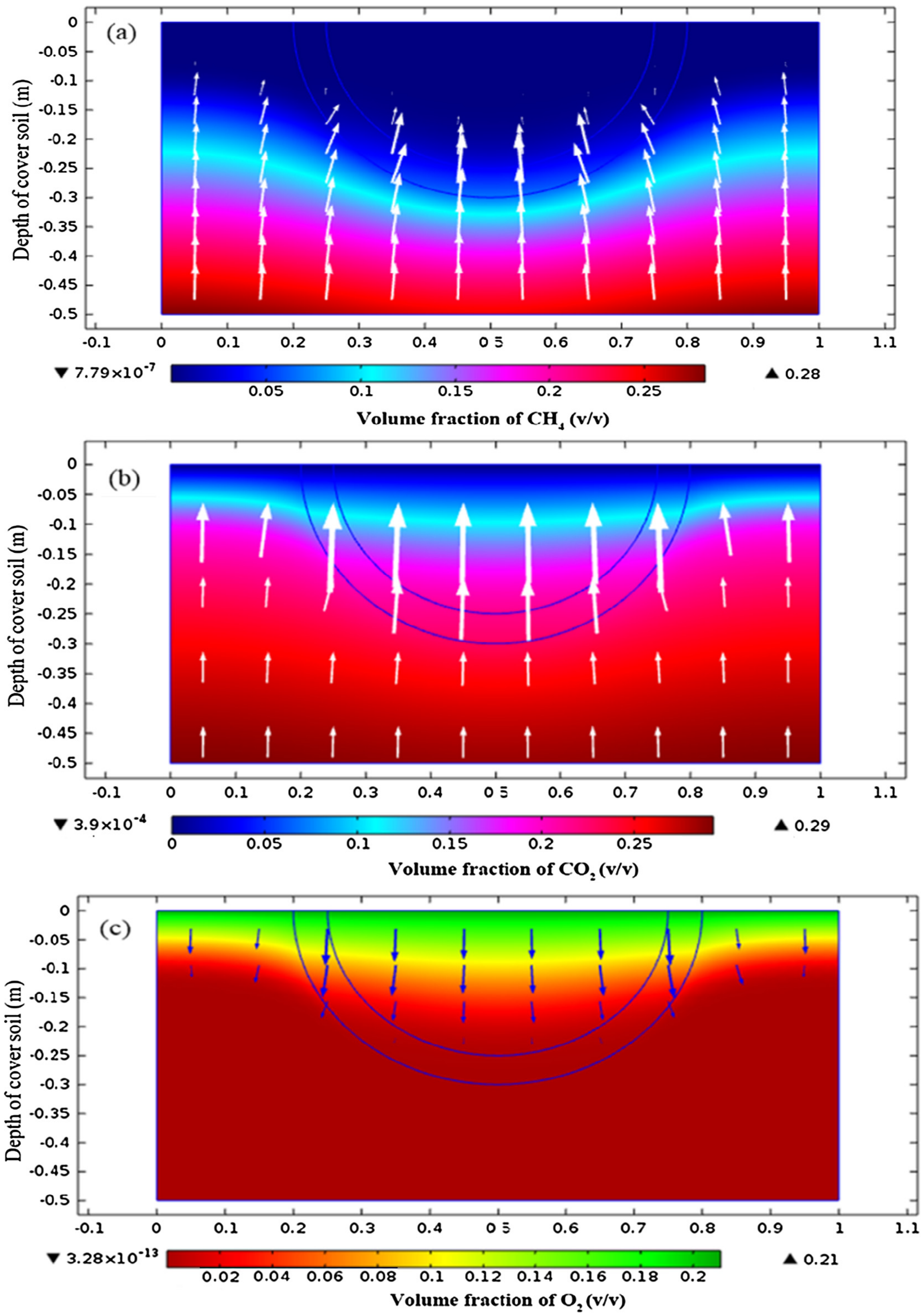

Fig. 5. Distribution of gas volume fraction (v/v) in the cover soil: (a) CH_4; (b) CO_2; (c) O_2. The arrows represent gas fluxes, and size of the gas flux increases with both length and thickness of the arrow.

3.3. Horizontal profile of CH_4 emission flux distribution in the cover soil

Horizontal profile of CH_4 and CO_2 distributions at depths of 0, 0.2 and 0.3 m are presented in Fig. 6 and Fig. 7. As CH_4 oxidation occurred at a depth of 0–0.2 m in the cover soil layer, the CH_4 emission flux decreased from the bottom to the upper cover soil, while CO_2 showed the opposite trend. The total CH_4 emission flux at the surface was significantly higher in vegetated areas than that in bare areas (Fig. 6(a)). The CH_4 emission flux ranged from 3×10^{-9} to 2×10^{-7} mol m^{-2} s^{-1} in vegetated areas, and from 10^{-10} to10^{-8} mol m^{-2} s^{-1} in bare areas. In total, CH_4 emission flux from vegetation areas was 1–2 orders of magnitude higher than bare areas. When CH_4 transport by vegetation was omitted (Fig. 6(a) red line), the CH_4 emission flux in vegetated areas was still higher

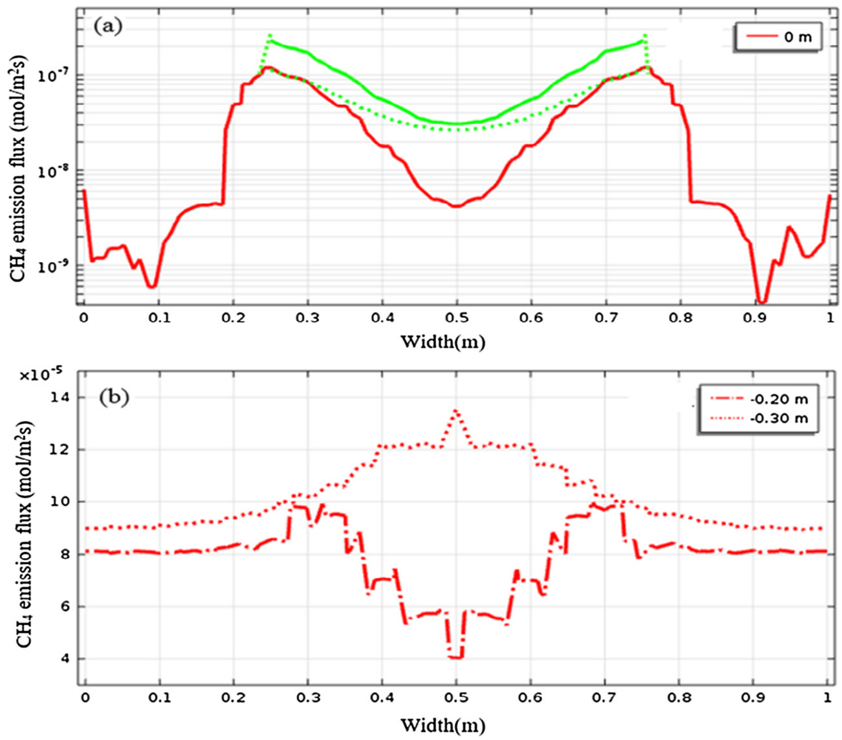

Fig. 6. CH_4 emission flux: (a) at the surface (the green dotted line is CH_4 emission flux from vegetation; the green line is the total CH_4 emission flux; the red line is CH_4 emission flux from cover soil); (b) at the depth of 0.20 m and 0.30 m. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

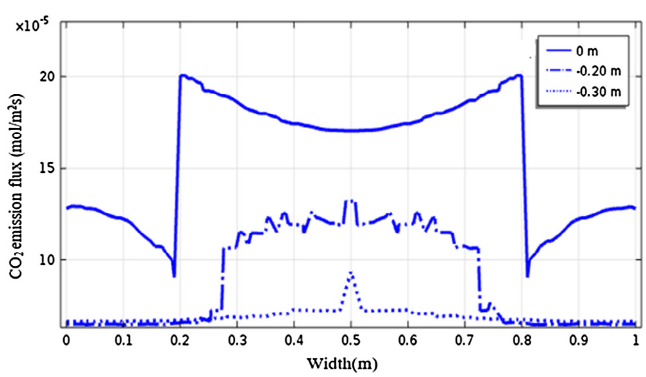

Fig. 7. CO_2 emission flux at different depths of cover soil.

than in bare areas as the higher soil porosity and diffusion coefficient caused by plant growth favored the CH_4 diffusion (Abichou et al., 2015). In vegetated areas, the highest CH_4 emission flux occurred at the interface of V1 and V2 at the depths of 0 and 0.2 m (2×10^{-7} mol m^{-2} s^{-1} and 9.92×10^{-5} mol m^{-2} s^{-1}, respectively). The pattern of CH_4 emission flux showed a concave shape in vegetated areas as the O_2 emission by the root system enhanced the CH_4 oxidation capacity (Fig. 6(a)). In the V1 area, the CH_4 emission flux by vegetation (Fig. 6(a) green dot line) was higher than that in bare areas (Fig. 6(a) red line). Vegetation hence represented the dominant CH_4 emission pathway. The CH_4 emission flux in bare areas was lower than in vegetated area at depths of 0.2–0.3 m due to the shallower O_2 penetrative depth in bare areas (Fig. 6(b)). The CH_4 emission flux at 0.3 m depth exhibited a completely contrasting pattern compared to the 0 and 0.2 m profiles, having a convex shape. It is the boundary of vegetation root influence areas and bare areas at the depth of 0.3 m, where the plant CH_4 transport takes place. Furthermore, it is also the diffusion lower boundary of O_2 emitting via root system. The suddenly decrease of O_2 concentration makes the CH_4 oxidation rate within a lower level. Thus the added plant CH_4 transport pathway and decreased CH_4 oxidation make the higher CH_4 emission rate at the depth of 0.3 m. Our simulated results concur with the theory that vegetation can mitigate CH_4 emission by enhanced CH_4 oxidation capacity, as well as accelerate CH_4 emission by enhanced CH_4 diffusion and transport by the vegetation.

3.4. CH_4 reaction rate distribution in cover soil

The CH_4 oxidation mostly occurred at a depth of 10 cm in the cover soil layer in bare areas. The range of CH_4 oxidation in vegetated areas, however, was much wider because of the enhanced O_2 diffusion capacity. When CH_4 transport by vegetation was omitted, the highest CH_4 reaction rate (0.0021 mol m^{-2} s^{-1}) occurred at the interface between bare and vegetated area (Fig. 8(a)). Enhanced soil porosity and CH_4 diffusion may cause the lower CH_4 oxidation rate in vegetated areas. However, the CH_4 oxidation rate in vegetated areas and bare areas decreased on the whole when CH_4 transport were included (Fig. 8(b)) as a result of the enhanced CH_4 transport capacity via vegetation. The highest CH_4 oxidation rate were only 0.0019 mol m^{-2} s^{-1} and 9.88×10^{-4} mol m^{-2} s^{-1} for vegetated and bare areas, respectively. The highest CH_4 reaction rate (0.00293 mol m^{-2} s^{-1}) was found in area V1 with a wide range due to the enhanced CH_4 transport via the roots of plants.

Above all, analytical results indicate that vegetation plays a key role in CH_4 transport, oxidation and emission. Vegetation can modify the O_2 distribution in cover soil by changing the properties of cover soil that enhance the oxidation capacity. At the same time, vegetation also transports CH_4 via the root system.

3.5. Model validation

The model was used to predict the CH_4 emission flux and oxidation percentage in two cells of Jiangcugou landfill covered with dif-

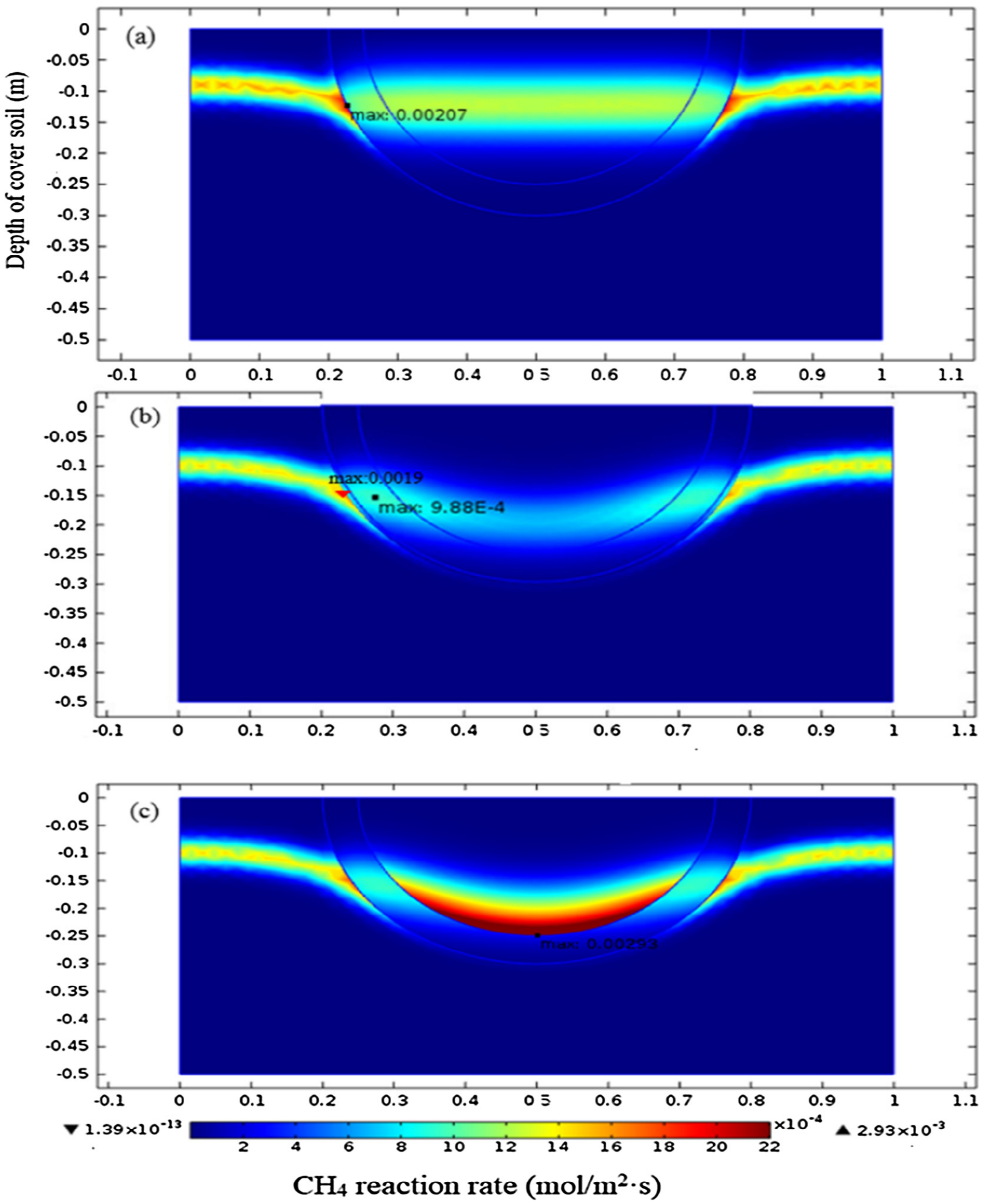

Fig. 8. The distribution of the CH_4 reaction rate in vegetated areas: (a) CH_4 oxidation rate without the direct CH_4 transport by the vegetation; (b) CH_4 oxidation rate with the direct CH_4 transport by the vegetation; (c) the total CH_4 reaction rate.

ferent vegetation. The predicted and filed measured values are shown in Table 4 based on the measured values of parameter (Table S2).

The predicted CH_4 emission flux by the model in bare areas X-B1 and X-B2 are 1.34×10^{-7} mol m^{-2} s^{-1} and 8.72×10^{-8} mol m^{-2} s^{-1} respectively, and the measured values are 1.30×10^{-7} mol m^{-2} s^{-1} and 1.07×10^{-7} mol m^{-2} s^{-1}. The relative deviations are 3.1% and -18.5% which indicates that our model can correctly predict CH_4 transportation, oxidation, and emission process in bare areas. However, the predicted value of CH_4 emission flux in the vegetated areas X-V1 is two orders of magnitude smaller than the measured value. Two reasons may explain this high deviation in area X-V1: (1) the quantity of O_2 release by plant roots is over estimated which causes a higher CH_4 oxidation ratio; (2) the improvement of CH_4 transportation efficiency causes by weather conditions is omitted, which lowers the CH_4 transportation flux via vegetation convection (Xin et al., 2016). The relative deviation of simulated value without the vegetation module is much smaller than with the vegetation module, and it underestimates the actual CH_4 emission flux which makes a lower contribution to the landfill greenhouse effect. The vegetation embedded model can hence modify the CH_4 emission, and improve the accuracy of the predicted value. Studies have shown that CH_4 emission is influenced by meteorological conditions such as the solar radiation, wind speed and temperature, as well as the cover soil properties (Abichou et al., 2011; Bohn et al., 2011; Reichenauer et al., 2011; Xin et al., 2016). The meteorological parameters in our model however are simplified. Correlation analysis between meteorological conditions and input parameters in the model are needed to further enhance and calibrate the model.

Table 4
Applications of this new model with field conditions.

Variable	Unit	X-B$_1$	X-V$_1$	X-B$_2$	X-V$_2$
CH_4 production value	mol m^{-2} s^{-1}	3.91×10^{-6}	3.42×10^{-6}	1.13×10^{-6}	3.42×10^{-6}
CH_4 measured value		1.30×10^{-7}	2.76×10^{-6}	1.07×10^{-7}	5.61×10^{-8}
CH_4 predicted value (with vegetation module)		1.34×10^{-7}	3.26×10^{-8}	8.72×10^{-8}	2.15×10^{-7}
CH_4 predicted value (without vegetation module)		–	6.12×10^{-9}	–	7.60×10^{-9}
Measured CH_4 oxidation percentage	%	97	19	91	98
Simulated CH_4 oxidation percentage	%	97	99	92	94

4. Conclusions

A novel simulation model was established in this study to incorporate the effect of vegetation on landfill CH_4 transport, oxidation and emission based on the interaction of vegetation and cover soil. The mechanisms of multicomponent diffusion and convection for landfill gas transport in the cover soil were included in this model and the plant-mediated transport was shown as the main route for CH_4 emissions. Vegetation should be considered as a priority factor in further developing landfill emission models. Limitations of this model is that the simulation model is a static model while parameters of vegetation and cover soil changed with meteorological conditions. Further modified studies with meteorological conditions are expected to improve the predicted accuracy of the model and facilitate the application of the model to modify landfill CH_4 mission inventory.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.wasman.2017.10.013.

References

Abichou, T., Chanton, J., Powelson, D., Fleiger, J., Escoriaza, S., Lei, Y., Stern, J., 2006. Methane flux and oxidation at two types of intermediate landfill covers. Waste Manage. 26, 1305–1312.
Abichou, T., Kormi, T., Yuan, L., Johnson, T., Francisco, E., 2015. Modeling the effects of vegetation on methane oxidation and emissions through soil landfill final covers across different climates. Waste Manage. 36, 230–240.
Abichou, T., Mahieu, K., Chanton, J., Romdhane, M., Mansouri, I., 2011. Scaling methane oxidation: from laboratory incubation experiments to landfill cover field conditions. Waste Manage. 31 (5), 978–986.
Abichou, T., Mahieu, K., Yuan, L., Chanton, J., Hater, G., 2009. Effects of compost biocovers on gas flow and methane oxidation in a landfill cover. Waste Manage. 29, 1595–1601.
Aydi, A., Abichou, T., Zairi, M., Sdiri, A., 2015. Assessment of electrical generation potential and viability of gas collection from fugitive emissions in a Tunisian landfill. Energy Strategy Reviews 8, 8–14.
Börjesson, G., Samuelsson, J., Chanton, J., 2007. Methane oxidation in Swedish landfills quantified with the stable carbon isotope technique in combination with an optical method for emitted methane. Environ. Sci. Technol. 41 (19), 6684–6690.
Babilotte, A., Lagier, T., Fiani, E., Taramini, V., 2010. Fugitive methane emissions from landfills: field comparison of five methods on a French landfill. J. Environ. Eng. 136 (8), 777–784.
Bogner, J.E., Spokas, K.A., Burton, E.A., 1997. Kinetics of methane oxidation in a landfill cover soil: temporal variations, a whole-landfill oxidation experiment, and modeling of net CH_4 emissions. Environ. Sci. Technol. 31 (9), 2504–2514.
Bohn, S., Brunke, P., Gebert, J., Jager, J., 2011. Improving the aeration of critical fine-grained landfill top cover material by vegetation to increase the microbial methane oxidation efficiency. Waste Manage. 31 (5), 854–863.
Chai, X., Tonjes, D.J., Mahajan, D., 2016. Methane emissions as energy reservoir: context, scope, causes and mitigation strategies. Prog. Energy Combust. Sci. 56, 33–70.
Colmer, T.D., 2002. Aerenchyma and an inducible barrier to radial oxygen loss facilitate root aeration in upland, paddy and deep-water rice (*Oryza sativa* L.). Ann. Bot. 91 (2), 301–309.
Colmer, T.D., 2003. Long-distance transportation of gases in plants: a perspective on internal aeration and radial oxygen loss from roots. Plant Cell Environ. 26 (1), 17–36.
De la Cruz, F.B., Green, R.B., Hater, G.R., Chanton, J.P., Thoma, E.D., Harvey, T.A., Barlaz, M.A., 2016. Comparison of field measurements to methane emissions models at a new landfill. Environ. Sci. Technol. 50, 9432–9441.
De Visscher, A., Thomas, D., Boeckx, P., Van Cleemput, O., 1999. Methane oxidation in simulated landfill cover soil environments. Environ. Sci. Technol. 33 (11), 1854–1859.
De Visscher, A., Van Cleemput, O., 2003. Simulation model for gas diffusion and methane oxidation in landfill cover soils. Waste Manage. 23 (7), 581–591.
EPA, 2013. Global Mitigation of Non-CO2 Greenhouse Gases: 2010–2030. United States Environmental Protection Agency (EPA), Washington DC.
Feng, S., Leung, A.K., Ng, C.W.W., Liu, H.W., 2017. Theoretical analysis of coupled effects of microbe and root architecture on methane oxidation in vegetated landfill covers. Sci. Total Environ. 599–600, 1954–1964.
Gabarro, J., Hernandez-Del Amo, E., Gich, F., Ruscalleda, M., Balaguer, M.D., Colprim, J., 2013. Nitrous oxide reduction genetic potential from the microbial community of an intermittently aerated partial nitritation SBR treating mature landfill leachate. Water Res. 47, 7066–7077.
Geck, C., Scharff, H., Pfeiffer, E.M., Gebert, J., 2016. Validation of a simple model to predict the performance of methane oxidation systems, using field data from a large scale biocover test field. Waste Manage. 56, 280–289.
Goldsmith, C.D., Chanton, J., Abichou, T., Swan, N., Green, R., Hater, G., 2012. Methane emissions from 20 landfills across the United States using vertical radial plume mapping. J. Air Waste Manage. Assoc. 62, 183–197.
Hilger, H.A., Liehr, S.K., Barlaz, M.A., 1999. Exopolysaccharide control of methane oxidation in landfill cover soil. J. Environ. Eng. Sci. 125 (12), 1113–1123.
IPCC, 2013. The final draft Report, dated 7 June 2013, of the Working Group I contribution to the IPCC 5th Assessment Report. In: Climate Change 2013: The Physical Science Basis.
Kirk, G., 1997. Changes in rice root architecture, porosity, and oxygen and proton release under phosphorus deficiency. New Phytol. 135, 191–200.
Knittel, K., Boetius, A., 2009. Anaerobic oxidation of methane: progress with an unknown process. Ann. Rev. Microbiol. 63, 311–334.
Kormi, T., Mhadhebi, S., Bel Hadj Ali, N., Abichou, T., Green, R., 2016. Estimation of fugitive landfill methane emissions using surface emission monitoring and Genetic Algorithms optimization. Waste Manage.
Krishna, R., Wesselingh, J.A., 1997. The maxwell-stefan approach to mass transfer. Chem. Eng. Sci. 52 (6), 861–911.
Li, T., Raivonen, M., Alekseychik, P., Aurela, M., Lohila, A., Zheng, X., Zhang, Q., Wang, G., Mammarella, I., Rinne, J., Yu, L., Xie, B., Vesala, T., Zhang, W., 2016. Importance of vegetation classes in modeling CH_4 emissions from boreal and subarctic wetlands in Finland. Sci. Total Environ. 572, 1111–1122.
Millington, R.J., Quirk, J.P., 1961. Permeability of porous solids. Trans. Faraday Soc. 57, 1200–1207.
Park, J.K., Kang, J.Y., Lee, N.H., 2016. Estimation of methane emission flux at landfill surface using laser methane detector: influence of gauge pressure. Waste Manage. Res. 34 (8), 784–792.
Park, J.R., Moon, S., Ahn, Y.M., Kim, J.Y., Nam, K., 2005. Determination of environmental factors influencing methane oxidation in a sandy landfill cover soil. Environ. Technol. 26, 93–102.
Rannaud, D., Cabral, A., Allaire, S.E., 2009. Modeling methane migration and oxidation in landfill cover materials with TOUGH2-LGM. Water Air Soil Pollut. 198 (1–4), 253–267.
Reichenauer, T.G., Watzinger, A., Riesing, J., Gerzabek, M.H., 2011. Impact of different plants on the gas profile of a landfill cover. Waste Manage. 31, 843–853.
Rusch, H., Rennenberg, H., 1998. Black alder (Alnus glutinosa (L.) Gaertn.) trees mediate methane and nitrous oxide emission from the soil to the atmosphere. Plant Soil 201 (1), 1–7.
Scheutz, C., Kjeldsen, P., Bogner, J.E., De Visscher, A., Gebert, J., Hilger, H.A., Huber-Humer, M., Spokas, K., 2009. Microbial methane oxidation processes and technologies for mitigation of landfill gas emissions. Waste Manage. Res.: J. Int. Solid Wastes Publ. Cleansing Assoc. ISWA 27 (5), 409–455.

Schuetz, C., Bogner, J., Chanton, J., Blake, D., Morcet, M., Kjeldsen, P., 2003. Comparative oxidation and net emissions of methane and selected non-methane organic compounds in landfill cover soils. Environ. Sci. Technol. 37 (22), 5150–5158.
Segers, R., Leffelaar, P.A., 2001. Modeling methane fluxes in wetlands with gas-transporting plants: 1. Single-root scale. J. Geophys. Res.: Atmos. 106, 3511–3528.
Spokas, K., Bogner, J., Chanton, J., 2011. A process-based inventory model for landfill CH4emissions inclusive of seasonal soil microclimate and CH4oxidation. J. Geophys. Res. 116.
Spokas, K., Bogner, J., Chanton, J., 2015. A process-based inventory model for landfill CH_4 emissions inclusive of seasonal soil microclimate and CH_4 oxidation. J. Geophys. Res. Biogeosci. 116 (G4), 4017.
Spokas, K., Bogner, J., Chanton, J.P., Morcet, M., Aran, C., Graff, C., Golvan, Y.M., Hebe, I., 2006. Methane mass balance at three landfill sites: what is the efficiency of capture by gas collection systems? Waste Manage. 26 (5), 516–525.
Stein, V.B., Hettiaratchi, J.P.A., Achari, G., 2001. Numerical model for biological oxidation and migration of methane in soils. Practice Periodical Hazard. Toxic Radioact. Waste Manage. 5 (4), 225–234.
Sun, Y., Yue, D., Li, R., Yang, T., Liu, S., 2015. Assessing the performance of gas collection systems in select Chinese landfills according to the LandGEM model: drawbacks and potential direction. Environ. Technol. 36, 2912–2918.
Taylor, D.M., Chow, F.K., Delkash, M., Imhoff, P.T., 2016. Numerical simulations to assess the tracer dilution method for measurement of landfill methane emissions. Waste Manage. 56, 298–309.
Vural, Y., Lin, M., Ingham, D.B., Pourkashanian, M., 2010. Comparison of the multicomponent mass transfer models for the prediction of the concentration overpotential for solid oxide fuel cell anodes. J. Power Sour. 195 (15), 4893–4904.
Webb, S.W., Pruess, K., 2003. The use of Fick's Law for modeling trace gas diffusion in porous media. Transp. Porous Media 51 (3), 327–341.
Wissuwa, M., Ae, N., 2001. Further characterization of two QTLs that increase phosphorus uptake of rice (*Oryza sativa* L.) under phosphorus deficiency. Plant Soil 237 (2), 275–286.
Xiaoli, C., Xin, Z., Ziyang, L., Shimaoka, T., Nakayama, H., Xianyan, C., Youcai, Z., 2011. Characteristics of vegetation and its relationship with landfill gas in closed landfill. Biomass and Bioenergy 35, 1295–1301.
Xin, D., Hao, Y., Shimaoka, T., Nakayama, H., Chai, X., 2016. Site specific diel methane emission mechanisms in landfills: a field validated process based on vegetation and climate factors. Environ. Pollut. 218, 673–680.
Xu, S., Jaffé, P.R., Mauzerall, D.L., 2007. A process-based model for methane emission from flooded rice paddy systems. Ecol. Model. 205, 475–491.
Yao, Y., Su, Y., Wu, Y., Liu, W., He, R., 2015. An analytical model for estimating the reduction of methane emission through landfill cover soils by methane oxidation. J. Hazard. Mater. 283, 871–879.
Zhang, D.Q., Tan, S.K., Gersberg, R.M., 2010. Municipal solid waste management in China: status, problems and challenges. J. Environ. Manage. 91 (8), 1623–1633.